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Analytical Correlations for Modeling the Laminar Flame Speed of Natural Gas Surrogate Mixtures

R. Amirante[1], E. Distaso[1*], P. Tamburrano[1] and R. D. Reitz[2]

[1] *Department of Mechanics, Mathematics and Management, Politecnico di Bari, Italy.*
[2] *Engine Research Center, University of Wisconsin-Madison, 1500 Engineering Drive, Madison, WI 53705, USA.*

Abstract

Natural gas is increasingly used as an alternative to petroleum fuels in internal combustion engines and industrial power plants because of its smaller environmental effects, as well as for economic reasons. Many applications, such as the spark-ignition engine simulations and the design of burners, require an accurate calculation of its laminar flame speed. Encouraging progress has been made in developing detailed chemical kinetic models for its prediction, but such models are still extremely complex and require significant computational effort. The laminar flame speed is an intrinsic property that is a function of the unburnt mixture composition, temperature, and pressure, therefore it is possible to develop analytical correlations based on experimental measurements, without losing accuracy, and that are more easily implemented in CFD codes than tabulated data.

The purpose of this study is to provide a simple, but accurate expression for modeling the laminar flame speed of natural gas as a function of its composition and over a wide range of operating conditions. In particular, a correlation valid for a natural gas ternary surrogate mixture of methane, ethane and propane is proposed. To achieve this aim, correlations for pure methane, as well as for binary methane/ethane and methane/propane mixtures were derived and combined to obtain a formulation suitable for different compositions of natural gas. It must be highlighted that some empirical correlations are already available in the literature, but they are usually based on a limited set of experimental measurements, thus they can fail outside the range in which they have been validated against experiments. In this study, measurements of laminar flame speeds obtained by several research teams are collected, compared, and critically analysed with the aim to develop more accurate empirical correlations. A comparison with available correlations in the literature shows the improvement in accuracy obtainable with the approach proposed in the present work.

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Keywords: Laminar flame speed, Epirical correlations, Natural Gas.

Corresponding Author: Tel. 3475910294,
e-mail address: elia.distaso@poliba.it

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ϕ	Equivalence ratio
p_0	Room pressure
p_u	Unburned mixture pressure
T_0	Room temperature
T_u	Unburned mixture temperature
S_L	Laminar flame speed
S_{L0}	Laminar flame speed at room conditions
α	Temperature influence exponent
β	Temperature influence exponent
B_m, B_2, ϕ_m and S_{u0}	Coefficients in Metghalchi et al.'s correlations
α_0, α_1 and α_2	Coefficients for exponent α
b_0, b_1 and b_2	Coefficients for exponent β
Z, W, η, ξ and σ	Coefficients in "Gülder's formulation" for pure compounds
ν, τ, ε and Ω	Coefficients in "Gülder's formulation" for fuel mixtures
χ	Volume fraction of other compounds in methane fuel mixtures

1. Introduction

Always more frequently, the design and optimization of many combustion applications, in which the flame propagation plays a primary role, relies on the use of computational simulations. And in such applications, a crucial aspect in determining the reliability of the numerical model is represented by the laminar flame speed estimation. Although encouraging progress has been made in developing detailed chemical kinetic models for its prediction, such models are still extremely complex and require significant computational effort for solving the mass, species and energy conservation equations coupled with chemistry [1,2]. In addition, they can fail outside the range in which they have been validated against experimental data, or if the grid resolution chosen for the simulation is not appropriate for the specific case [3]. This is because flame propagation usually involves time and spatial scales which cannot be typically captured with practical finite volume methods [4].

Thus, analytical correlations of the laminar flame speeds as a function of equivalence ratio, pressure and temperature are preferred in practical simulations. Moreover, they are more easily implemented in CFD codes than tabulated data. Such an approach is originated from the consideration that the laminar flame speed is uniquely defined, for a given fuel, once the unburned mixture composition, temperature, and pressure are known.

In the last sixty years, various forms of empirical and semi-empirical functional relationships have been proposed for the laminar burning velocity [5,6]. The simplest alternative and the most widely used form of the wholly empirical correlation is the so-called "power law" formula, adopted by many Investigators [7–10]:

$$S_L(\phi, T_u, p_u) = S_{L0}\left(\frac{T_u}{T_0}\right)^{\alpha}\left(\frac{p_u}{p_0}\right)^{\beta}, \quad (1)$$

where S_{L0} is the velocity, for a given equivalence ratio ϕ, measured at room conditions, namely at $T_u = T_0$ and $p_u = p_0$, and α and β are constants or mixture strength-dependent terms.

An expression for the term $S_{L0}(\phi)$ in Equation (1) was proposed by Gülder [6]:

$$S_{L0}(\phi) = Z\,W\,\phi^{\eta}\,e^{-\xi(\phi-\sigma)^2}, \quad (2)$$

where W, η and ξ are constants for a given fuel, and $Z = 1$ for single constituent fuels.

The usage of natural gas and the interest towards it have risen because of its smaller environmental effects compared to conventional petroleum fuels and its improved availability due to shale gas [11–16]. Natural gas is also an attracting alternative for transport due to its potentially smaller gaseous and particulate emissions [13,15,17]. In the engine field new combustion techniques [18,19] are related to the use of natural gas, as well as their control strategies [20,21]. It appears crucial developing precise and reliable correlations for estimating its laminar flame speed. However, natural gas is a mixture of various hydrocarbon molecules and their volume fraction can considerably vary with geographical source, time of year, and treatments applied during production or transportation [22]. It has been demonstrated that a simple variation of the Z parameter in the correlation proposed by Gülder [6] is not sufficient to capture the effects of variation of natural gas composition on its laminar flame speed.

Fuel	$W\ [cm/s]$	η	ξ	σ	ν	τ	Ω
CH_4/ C_2H_6	38.638	-0.15	6.2706	1.1	0.2103	0.545	-0.0191
CH_4/ C_3H_8					0.2129	0.8312	-0.0439

Table 2. Coefficients proposed by Liao et al. [14] for exponents α and β in Equation (5) for a Chinese Natural Gas.

Fuel	a_2	a_1	a_0	b_2	b_1	b_0
Natural Gas	5.7500	12.150	7.9800	0.9250	2.0000	1.4730

Recently, Dirrenberger et al. [10] used the following modified form of Equation (2), proposed by Coppens et al. [23] in 2007, for calculating the laminar flame speed of binary methane/ethane and methane/propane mixtures, as follows:

$$S_{L0}(\phi,\chi) = (1+\nu\chi^{\tau})\, W\, \phi^{\eta}\, e^{-\xi(\phi-\sigma-\Omega\chi)^2}. \tag{3}$$

The term Z, present in Gülder's formulation (Equation (1)), assumes the value $(1+\nu\chi^{\tau})$ to take into account the presence of other compounds in methane. χ is the amount of the other gas in the fuel mixture. The additional term $\Omega\chi$ in the exponent, allows to reproduce the shift of the maximum of the laminar flame velocity's dependence with the additional gas concentration. When χ is zero, the original Gülder's formulation for pure compounds is obtained. The coefficients derived from the experimental data interpolation are reported in Table 1.

In the same work, Dirrenberger et al. [10] extended the formulation to ternary mixture by combining the expressions formulated by Coppens et al. [23] for binary mixtures. They obtained a relationship valid for a natural gas surrogate mixture of methane, ethane and propane:

$$S_{L0}(\phi,\chi_1,\chi_2) = (1+\nu_1\chi_1^{\tau_1})(1+\nu_2\chi_2^{\tau_2})\, W\, \phi^{\eta}\, e^{-\xi(\phi-\sigma-\Omega_1\chi_1-\Omega_2\chi_2)^2}, \tag{4}$$

where the subscript 1 refers to parameters calculated for one component, i.e. ethane, and subscript 2 to the other one, i.e., propane. Once again, if $\chi_1 = \chi_2 = 0$ the correlation for pure fuels is obtained. If either $\chi_1 = 0$ or $\chi_2 = 0$, then the previous binary mixtures formulation is derived.

The only study available in the literature in which the dependence of the exponents α and β upon the equivalence ratio was investigated in the case of natural gas, is by Liao et al. [9]. In particular, they studied a Chinese Natural Gas (from the north of Shannxi Province) and they proposed the following second-order polynomial form, with the coefficients reported in Table 2:

$$\begin{aligned}\alpha(\phi) &= a_2\phi^2 - a_1\phi + a_0\\ \beta(\phi) &= -b_2\phi^2 + b_1\phi - b_0,\end{aligned} \tag{5}$$

The aim of the present study, is to provide improved empirical correlations for calculating the laminar flame speed as functions of equivalence ratio and unburned mixture temperature and pressure of binary mixtures, of binary mixture of methane/ethane and methane/propane as well as for ternary mixtures composing various natural gases, starting from those proposed by Coppens et al. [23] and Dirrenberger et al. [10], respectively.

2. Results and discussions

The correlations proposed in this work have the "power law" form of Equation (1), with $p_0 = 1\ atm$ and $T_0 = 298\ K$. The exponents α and β were considered to be functions of the mixture strength ϕ and the second-order polynomial fitting proposed by Liao et al. [9] (Equation (5)) is considered.

In order that an analytical formulation (as well as a chemical kinetics mechanism) can be considered reliable for many possible conditions, it must be validated against a large body of data. Thus, in this work, experimental measurements of laminar flame speeds, carried out by several workers are compared and critically evaluated. Results for the considered studies are listed in tables, together with the method that was used, the range of the equivalence ratios, pressures and temperatures that were explored, and the fuels that were considered in the specific study.

An iterative reweighted least squares algorithm was performed for the parameter estimation. In particular, a nonlinear regression using ordinary least squares, coupled with the Levenberg-Marquardt Algorithm [24], was adopted.

2.1. Correlation for Methane/propane and methane/ethane binary mixtures

A study of binary mixtures of methane with ethane and propane is crucial for developing empirical correlations able to reproduce the laminar flame speed of different types of natural gas, since the methane fraction can vary between 55.8% and 98.1%, ethane can vary between 0.5% and 13.3%, and propane can vary between 0% and 23.7% [22].

Dirrenberger et al. [10] proposed a modified version of Gülder's expression (Equation (3)) to take into account the presence of another compound with methane. They found that their correlation reproduced well the experimental results for lean and rich mixtures, but overestimated flame velocities near stoichiometry. This because their modifications considered only the influence on the peak amplitude and position. In fact, as it is inferable from their experimental data, the lean and the rich side are more sensitive to the addition of another compound. Therefore, in order to take into account such behaviour an improved formulation of Equation (3) was studied. In particular, the coefficient η in Equation (3) has been multiplied by the term $(1-\chi)^{\varepsilon}$, resulting in the following expression:

$$S_{L0}(\phi,\chi) = (1+\nu\chi^{\tau})\, W\, \phi^{\eta(1-\chi)^{\varepsilon}}\, e^{-\xi(\phi-\sigma-\Omega\chi)^2}. \quad (6)$$

The introduction of the new term allows to reproduce the fact that the equivalence ratio has different influence on different mixture compositions, and it has the strongest effects in conditions far from the stoichiometric one. The coefficients ν, τ, ε and Ω derived in this study for methane/ethane and methane/propane mixtures are reported in Table 4. The terms W, η, ξ and σ, reported in Table 3, refer to pure methane and were derived and validate in a previous study [5].

The works that were considered for the analysis are reported in Table 5. Figure 1 shows the results for different fractions of ethane in methane, while Figure 2 refers to methane/propane mixtures. From Figure 1(a) and Figure 2(a) it is seen that the proposed correlation reproduces the experimental trends better than the formulation proposed by Dirrenberger et al. [10], for all equivalence ratios considered. It captures the greater sensitivity to the addition of other compounds in methane for lean and rich mixtures.

Figure 1(b), (c) and (d) offer a comparison with other experimental measurements for methane/ethane, and Figure 2(b) compares methane/propane mixtures. The overall agreement can be considered satisfactory.

2.2. Correlation for Natural Gas

Dirrenberger et al. [10] proposed a correlation valid for a natural gas surrogate mixture of methane, ethane and propane, which was obtained by combining the expressions derived for binary methane/ethane and methane/propane mixtures. The same approach has been adopted in this study, resulting in the following expression:

$$S_{L0}(\phi,\chi_1,\chi_2) = (1+\nu\chi_1^{\tau_1})(1+\nu\chi_2^{\tau_2})\, W\, \phi^{\eta(1-\chi_1)^{\varepsilon_1}(1-\chi_2)^{\varepsilon_2}}\, e^{-\xi(\phi-\sigma-\Omega_1\chi_1-\Omega_2\chi_2)^2}, \quad (7)$$

Table 3. Coefficients for Equation (2) derived in a previous study [6] for methane.

Fuel	Z	$W\ [cm/s]$	η	ξ	σ
CH_4	1	38.85	-0.20	6.45	1.08

Table 4. Coefficients of Equation (6) for binary mixtures.

Fuel	ν	τ	ε	Ω
CH_4/ C_2H_6	0.20	1.50	0.95	0.09
CH_4/ C_3H_8	0.10	1.50	1.30	0.20

Table 5. Literature considered for methane/ethane and methane/propane mixtures.

Ref.	Authors	year	Fuels	Phi	T_u [K]	p_u [atm]	Method
[25]	Kishore	2008	Methane, Ethane, Methane/Ethane mixtures	0.7-1.3	307	1	Heat flux
[26]	Lowry	2011	Methane, Ethane, Propane Methane/Ethane, Methane/Propane	0.7-1.3	298	1-10	Constant-volume vessel with schlieren optical setup
[10]	Dirrenberger	2011	Methane, Ethane, Propane, n-Butane, Methane/Ethane, Methane/Propane, Natural Gas	0.6-2.1	298	1	Heat flux method with flame adiabatic burner / Correlations

in which the terms W, η, ξ and σ refer to pure methane (Table 3), while the coefficients ν, τ, ε and Ω for ethane and propane are reported in Table 4. The works considered are listed in Table 6.

Dirrenberger et al. [10] studied three surrogate mixtures with compositions close to those of three representative natural gases: Indonesia, Abu Dhabi and Pittsburgh, Table 7. shows the exact composition of these natural gases. In such study, they were represented by the following mixtures: 90% CH_4, 6% C_2H_6, and 4% C_3H_8 Indonesia, 82% CH_4, 16% C_2H_6, and 2% C_3H_8 Abu Dhabi and 85% CH_4 and 15% C_2H_6 Pittsburgh. The results for each natural gas are reported in Figure 3, together with a comparison with the empirical correlation proposed by Dirrenberger et al. [10] (Equation (4)). The dependence upon the equivalence ratio and the fuel composition is well captured by the present proposed correlation and it shows better agreement, especially near stoichiometry.

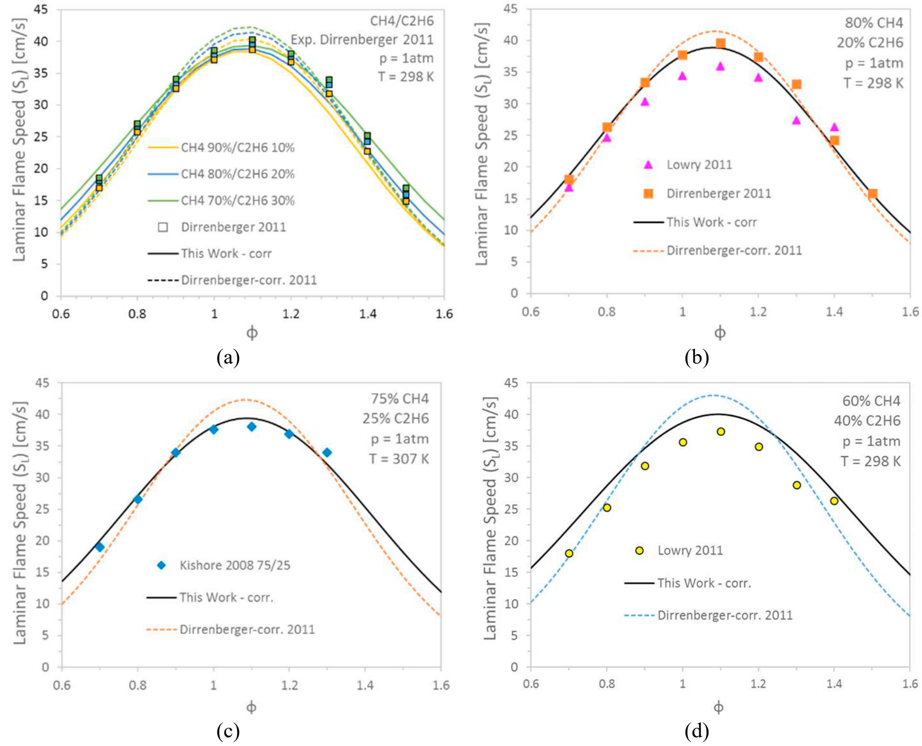


Figure 1. Laminar flame speed of methane/ethane mixtures at room conditions, considering different ethane content in methane. Marks: experimental data; dashed lines: correlations proposed by Dirrenberger et al. [10]; solid line: empirical correlation proposed in this work.

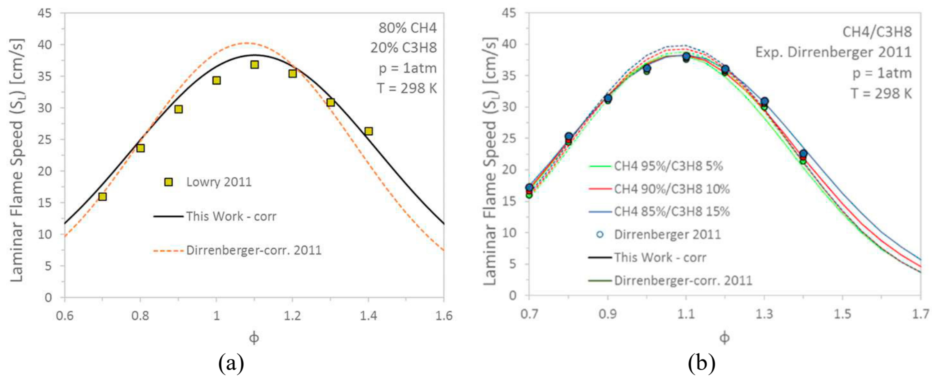


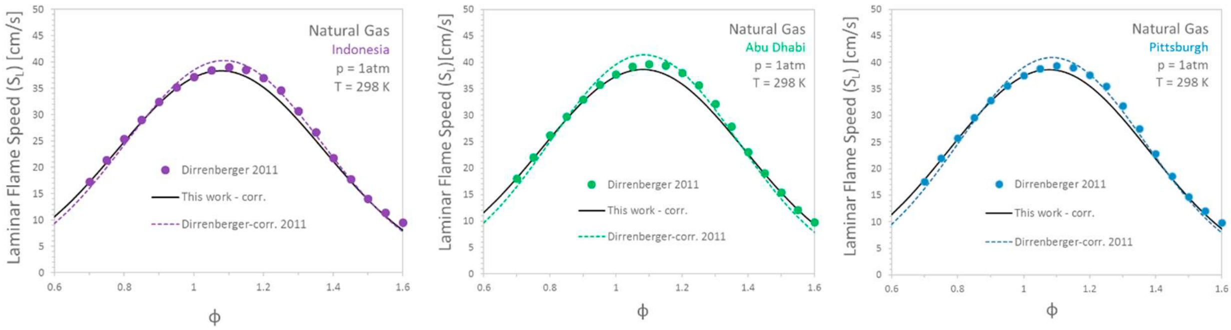
Figure 2. Laminar flame speed of methane/propane mixtures at room conditions, considering different ethane content in methane. Marks: experimental data; dashed lines: correlations available in literature; solid line: empirical correlation proposed in this work.

Table 6. Literature considered for natural gas.

Ref.	Authors	year	Fuels	Phi	T_u [K]	p_u [atm]	Method
[9]	Liao	2004	Shannxi Natural Gas	0.6-1.4	300-400	0.5-1.5	Spherical bomb
[27]	Bourque	2010	Methane, Natural gas	0.7-1.3	298	1-4	Cylindrical bomb with Schlieren setup
[10]	Dirrenberger	2011	Methane, Ethane, Propane, Butane, Methane/Ethane, Methane/Propane, Natural Gas	0.6-2.1	298	1	Heat flux method with flame adiabatic burner / Correlations

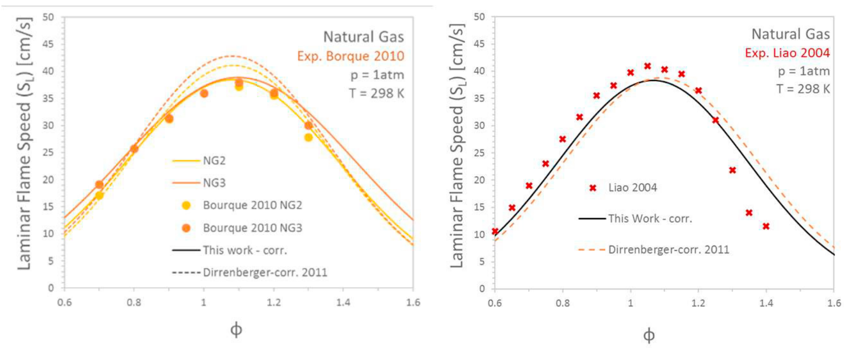
Table 7. Composition of different natural gases (% Volume) considered.

Ref.	Authors	year	Fuels	CH_4	C_2H_6	C_3H_8	i-C_4H_{10}	n- C_4H_{10}	i-C_5H_{12}	n-C_5H_{12}	CO_2	N_2	others
[10]	Dirrenberger	2011	Indonesia	89.91	5.44	3.16	1	0.75	0.03	-	-	0.04	-
			Abu Dhabi	82.07	15.86	1.89	-	0.06	-	-	-	0.05	-
			Pittsburgh	85.00	14.0	-	-	-	-	-	-	1.00	-
[27]	Bourque	2010	NG2	81.25	10.0	5.0	-	2.50	-	1.25	-	-	-
			NG3	62.50	20.0	10.0	-	5.00	-	2.50	-	-	-
[9]	Liao	2004	Shannxi	96.16	1.096	-	-	-	-	-			2.74



(a) (b) (c)

Figure 3. Laminar flame speeds of different natural gases at room conditions. Marks: experimental data; dashed lines: correlations proposed by Dirrenberger et al. [10]; solid line: empirical correlation proposed in this work.



(a) (b)

Figure 4. Laminar flame speed of different natural gases measured by Bourque et al. [27] (a) and Liao et al. [9] at room conditions. Marks: experimental data; dashed lines: correlations proposed by Dirrenberger et al. [10]; solid line: empirical correlation proposed in this work.

Table 7 also reports the composition of the two natural gas mixtures that were the focus of the study by Bourque et al. [27]. they were represented by the following mixtures: 85% CH_4, 10% C_2H_6, and 5% C_3H_8 for NG2, 70% CH_4, 20% C_2H_6, and 10% C_3H_8 for NG3. The results for each natural gas are reported in Figure 4(a). The Dirrenberger et al. correlation [10] overestimates the maximum flame speed, but the correlation proposed in this study shows an overall better agreement with the experiments. In addition, it is able to capture the influence of the composition far from stoichiometry.

Figure 4(b) reports results for the natural gas investigated by Liao et al. [9]. Its composition is reported in Table 7 as well. It was represented by considering 98.9% CH_4 and 1.1% C_2H_6. The values of the laminar flame speed reported in this study appear to be higher than those of the cases previously investigated, even though the natural gas was composed almost exclusively of methane. These experimental measurements report values that are higher than those presented previously for pure methane. Therefore, it is hard to judge the results shown in Figure 4(b).

Very few works have investigated the influence of initial pressure and temperature on natural gas laminar flame speed. Figure 5(a) shows results for different initial pressures and Figure 5(b) different initial temperatures. Only the stochiometric case has been investigated. The derived values for the coefficients of exponents α and β in Equation (5) are listed in Table 2. Liao et al. [9] investigated the temperature influence (the coefficients of exponent α are the same proposed by Liao et al. [9]).

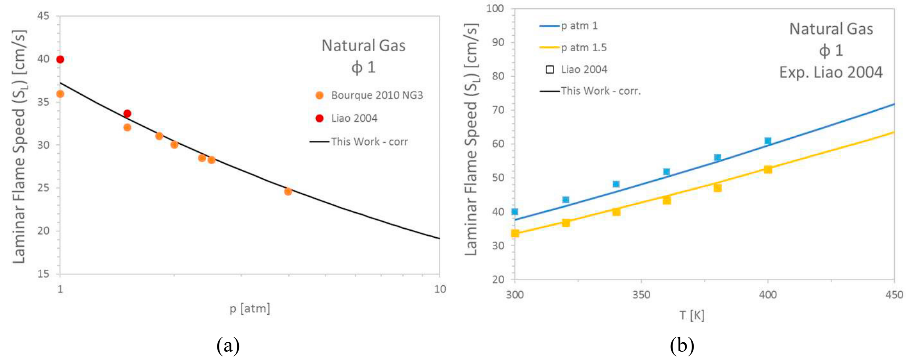


Figure 5. Initial pressure (a) and temperature (b) influence on natural gas laminar flame speed at stoichiometric conditions. Marks: experimental data; solid lines: empirical correlation proposed in this work.

3. Conclusions

The present study provides simple and reliable expressions that allow laminar flame speed calculations of binary mixtures of methane/ethane and methane/propane, as well as different type of natural gasses. Empirical correlations available in literature generally are usually not able to give good agreement with recent experimental data. This because many of them are based on a single set of measurements and fail outside the considered experimental range. Therefore, measurements of laminar flame speeds in literature were collected and used to develop more accurate and reliable empirical correlations.

The correlations proposed in this work have the "power law" form (Equation (1)), with $p_0 = 1\ atm$ and $T_0 = 298\ K$. $S_{L0}(\phi)$ term is represented using "Gülder's exponential formulation" (Equation (2)), while the exponents α and β were functions of mixture strength ϕ and a second-order polynomial fitting was considered (Equation (5)).

For binary and ternary mixtures, it was shown that the influence that the amount of the secondary compounds has on the mixture laminar flame speed is different at different equivalence ratios which has not been considered in previous formulations. Therefore, a modified expression for the term $S_{L0}(\phi)$ was proposed (Equation (6)), and better overall agreements with all the experimental data was obtained.

An improved formulation was developed for calculating the laminar flame speed of natural gas (Equation (7)), which was modelled as a ternary mixture of methane, ethane and propane. Comparisons with experimental data on natural gases having different compositions confirmed the obtained improvements.

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